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MEASUREMENTS OF ENRICHED URANIUM**

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NEUTRON AND Γ PULSE SHAPE DISCRIMINATION IN A LIQUID SCINTILLATOR COUNTER FOR NEUTRON MULTIPLICITY MEASUREMENTS OF ENRICHED

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Abstract

A new neutron multiplicity counter is being developed which utilizes the fast response of liquid scintillator (NE-213) detectors. Current uranium coincidence counting methods rely on the assay samples to conform to the calibration standards with respect to the sample uniformity, geometry, material type, etc. There exists a wide range of material throughout the DOE complex where these attributes are non-standard or unknown. A neutron counter with short die-away time makes possible the measurement of higher order coincidences. This information can be used to more accurately assay many of the problem items in the inventory. In addition, such a counter would allow for rapid inventory measurements of all forms of uranium. Liquid scintillator detectors also allow for energy discrimination between interrogation source neutrons and fission neutrons, allowing for even greater assay sensitivity.

Liquid scintillator detectors are sensitive to γ and neutron radiation. Differences in the timing of scintillation light produced in γ -ray and neutron interactions allows for separation of these events using pulse shape discrimination (PSD). PMT pulses resulting from neutron and γ interactions in the Liquid scintillator are read in using a fast waveform digitizer with a 1 GS/s sampling rate. The pulse shapes are then compared to γ and neutron pulse templates and a χ^2 comparison determines the species. Integrated rise time can also be used to discriminate between the γ and neutron pulses. The results of these studies are presented.

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I. Introduction

Active multiplicity counting has been under development at Los Alamos National Laboratory for several years.^{1,2,3,4,5} Neutron multiplicity counters (NMC) based on ^3He systems are a valuable tool for the nondestructive assay (NDA) of special nuclear material. However, there exists a wide range of materials throughout the DOE complex that still can't be measured (remote-handled waste, spent fuel, very impure Pu, bulk highly enriched uranium). A fast neutron counter overcomes many of the difficulties present in measuring samples that do not conform to available standards. A shorter die-away time reduces the accidental coincidence rate and enables us to measure higher moments of the multiplicity distribution with greater precision in a short period of time. Thus, sample properties such as self-multiplication and (α, n) reaction rate can be accurately determined instead of relying on previous knowledge of a given matrix.

These advantages of a fast neutron counter are even more pronounced in the assay of highly enriched uranium (HEU) for a couple of reasons. The technique of uranium

assay by coincidence counting relies heavily on the calibration standards being representative of the measurement items. The technique of active multiplicity counting has been seen to be insensitive to the material type and geometry of the calibration standard used. The drawback of active multiplicity counting is that it requires an accurate measurement of the triples count rate, which can only be measured in a reasonable time with a fast neutron counter. In addition, for uranium assay the AmLi activation neutrons contribute to the accidental coincidence rate. Unlike ^3He -based systems in which neutrons are moderated and then detected as thermal neutrons, a fast neutron counter retains the spectral information. This allows us to set a threshold, which removes a large part of the low energy activation neutrons and allows for the measurement of the coincidence count rates to a much better precision in a shorter period of time.

Figure-of-merit (FOM) simulation codes predict that an order of magnitude improvement in facility throughput or range of sample size can be achieved with coincidence resolving times on the order of tens of nanoseconds. The relative assay error

is roughly proportional to $\frac{\sqrt{\tau}}{\epsilon \sqrt{t}}$ (where τ is the die-away time, ϵ is the efficiency and t is

the measurement time). This improvement will be achieved for both passive and active multiplicity counters, but it is most needed for active multiplicity counting of the huge inventories of HEU metals, oxides, scrap, and residue items present in DOE facilities. Liquid scintillator detectors are sensitive to γ and neutron radiation.

Unlike traditional ^3He counters, NE-213 is sensitive to both γ -rays and neutrons. Differences in the timing of scintillation light produced in γ -ray and neutron interactions, however, allows for separation of these events using pulse shape discrimination (PSD). Historically, analogue electronics was used to perform pulse shape discrimination (PSD) to separate γ and neutron interactions. Recently, the availability of fast waveform digitizers has made possible alternate methods for PSD.

A fast waveform digitizer with a 1 GS/s sampling rate is used for data acquisition and software has been developed to interface with the digitizer. PSD can be performed using only the first 250-500 ns of the pulse, thus allowing for a short gate window and reducing the sensitivity to random coincidences. In addition, this data acquisition technique preserves the full information about each pulse. This means that a number of signal processing algorithms can be used for PSD, including some that are not possible in analog electronics. Presented here are preliminary studies in the development of these PSD methods.

II. Data Acquisition

The liquid scintillator module used in this study consists of a volume (5 inches diameter by 8 inches long) of NE-213 liquid scintillator optically coupled to a 5-inch photomultiplier tube (PMT). A possible well counter design consisting of 12 such modules is shown in Figure 1. A possible well counter with 12 modules, each consisting of a NE-213 volume optically coupled to a PMT.. To study the pulse shape discrimination, a single module was used. Several sources (^{252}Cf , AmLi, and ^{137}Cs) were placed in a lead cylinder near the liquid scintillator module (see Figure 2). PMT pulses

were fed into an Acqiris DC271 cPCI digitizer module¹ with a 1GS/sec sampling rate (see Figure 2). A duplicate PMT pulse is fed through a discriminator and used as a trigger. For each trigger, digitized pulse data is written to disk for a 500ns time window.

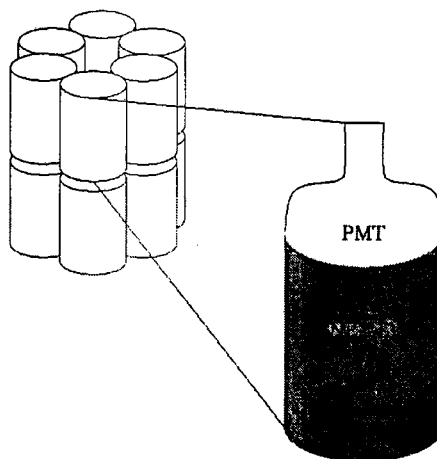


Figure 1. A possible well counter with 12 modules, each consisting of a NE-213 volume optically coupled to a PMT.

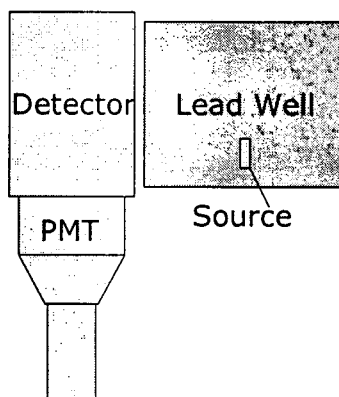


Figure 2. Experimental setup. The PMT + NE213 module is oriented with the NE213 volume above the PMT. Sources are placed in the center of a lead cylinder placed next to the NE213 volume. The cylinder has a 1 inch wall thickness and 7 and 9 inch inner and outer diameter respectively.

The raw digitized pulses are stored on disk and can then be analyzed offline. In future, PSD software will be integrated into the data acquisition software to reduce the disk space needs and speed up processing time. The offline analysis software searches for the maximum of the peak position and defines the pulse from 50ns before to 150ns after the pulse maximum. Then, the various PSD algorithms are applied to this data.

¹ 3 channels are used to increase the resolution of the 8 bit ADC associated with each channel. The first channel ranges from 0.0-0.5 Volts, the second 0.0-1.0 V and the third 0.0-2.0 V. The 3 channels are later combined in reconstruction software.

III. PSD Algorithms

Both γ -rays and neutrons entering the NE-213 volume will produce light. Luckily, these interactions result in different PMT pulse shapes, so that the γ events can be separated from the neutron events. The PMT charge pulse resulting from a neutron interaction will have a much longer tail than the pulse resulting from a γ -ray interaction (see Figure 3) and the integral of the neutron pulse will rise to its total value less sharply than the γ pulse.

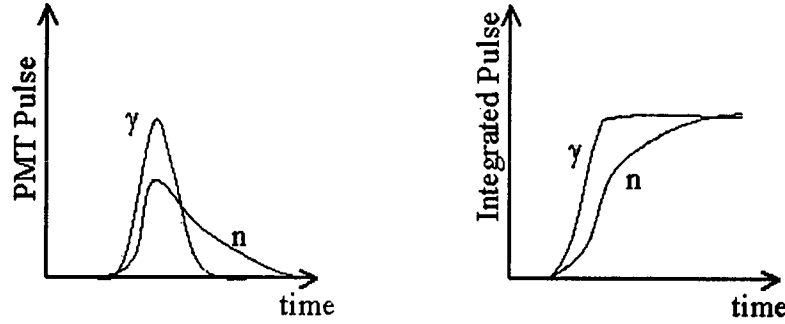


Figure 3. PMT charge pulse and integrated pulse versus time for γ and neutron events.

We have investigated 2 PSD methods. The first method, χ^2 , compares the PMT pulses resulting from neutron and γ interactions in the liquid scintillator to γ and neutron pulse templates, and a χ^2 comparison determines the species. The initial template pulses are defined using γ source data and Monte Carlo neutron pulse data. The χ^2 method is then applied to fission source data with these templates and a second set of template pulses can be defined by the sorted pulses of the first iteration. Figure 4 shows the neutron and γ pulse templates. The second method, Combined Algorithms (CA), utilizes a weighting scheme to combine the following 4 PSD algorithms (see Figure 5):

1. The Integral Rise Time (IR) algorithm compares the times at which a pulse reaches 80% of its total integrated value. Neutrons have a larger IR value
2. The Decay Time (DT) algorithm compares the times at which a pulse decays to 12% of the peak maximum. Neutrons have a larger DT value
3. The Short Integral (SI) algorithm compares the ratio of a short integration time window (70ns) to the total integration (200ns). Neutrons have a smaller SI value
4. The Pulse Height (PH) algorithm compares the ratio of pulse height to pulse width. Neutrons have a smaller PH value.

The weights are determined empirically to give good numbers for efficiency and γ contamination. Above 300 KeV electron energy (see discussion of energy calibration below), the SI algorithm is used solely.

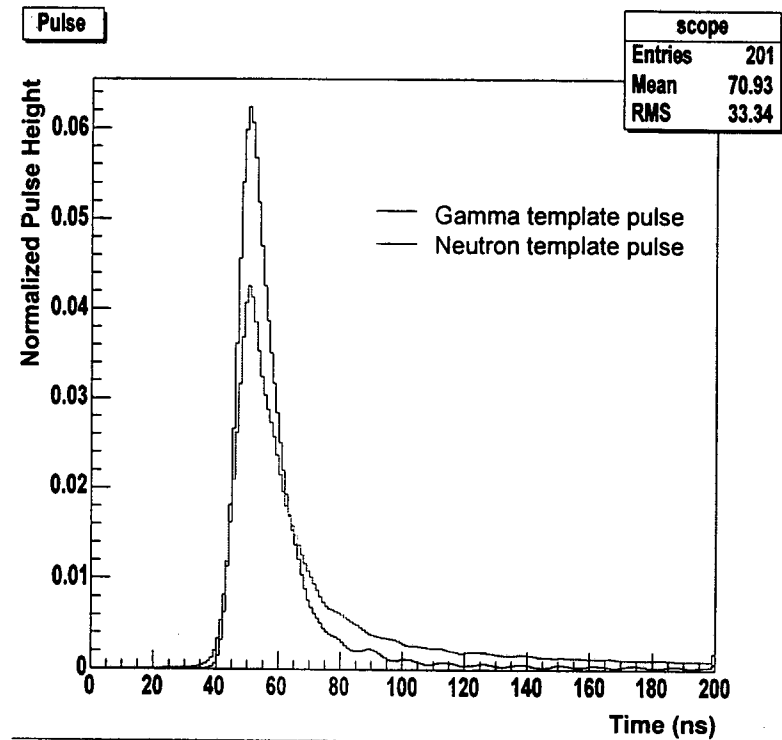


Figure 4 γ and neutron template pulses from ^{252}Cf data. Area's under the two curves are normalized to 1.

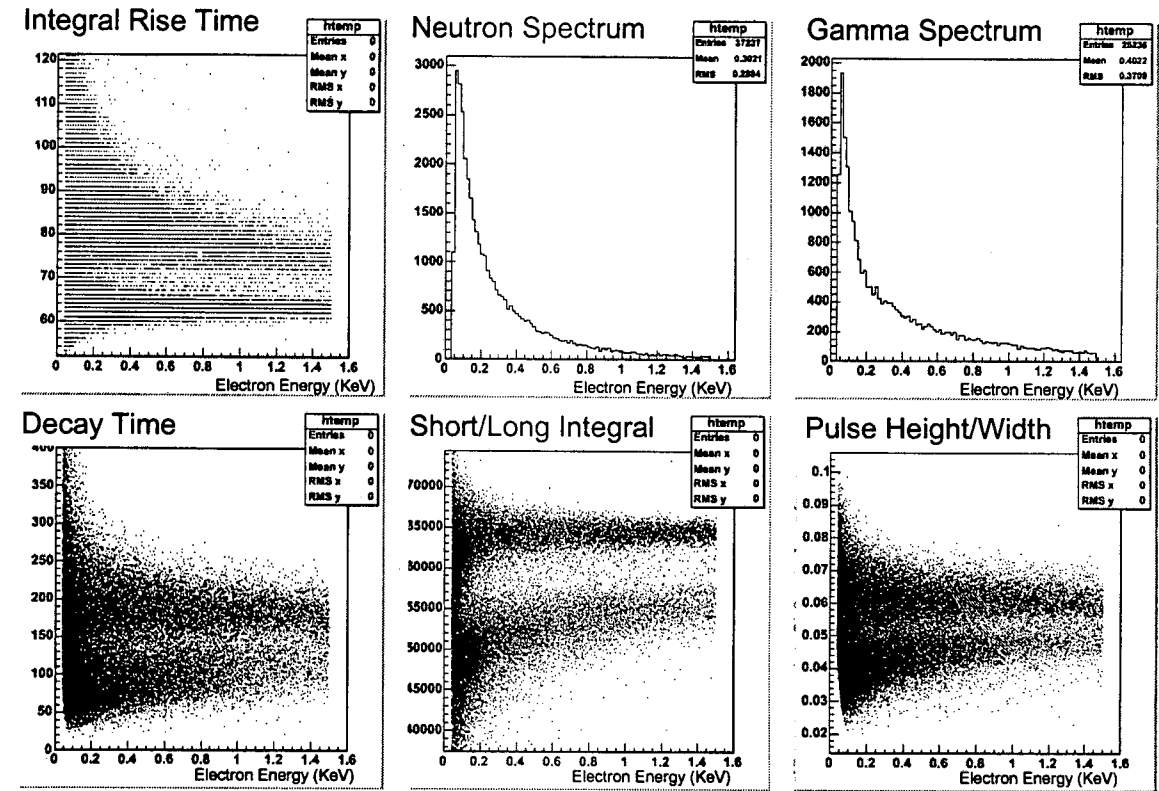


Figure 5 The 4 PSD algorithms in the CA method. In Black are events determined to be neutrons and in red are γ s.

III. Efficiency and γ Contamination

The efficiency and γ contamination are measured for several energy thresholds. Energy calibration is performed using ^{137}Cs γ source data. A theoretical γ Compton edge spectrum smeared by a fixed detector resolution is fit to data (see Figure 6). This gives the electron recoil energy, E_e , calibration. This is translated to proton recoil energy, E_p , through the relation, $E_e (\text{MeV}) = 0.215 E_p + 0.028 E_p^2$ [6].

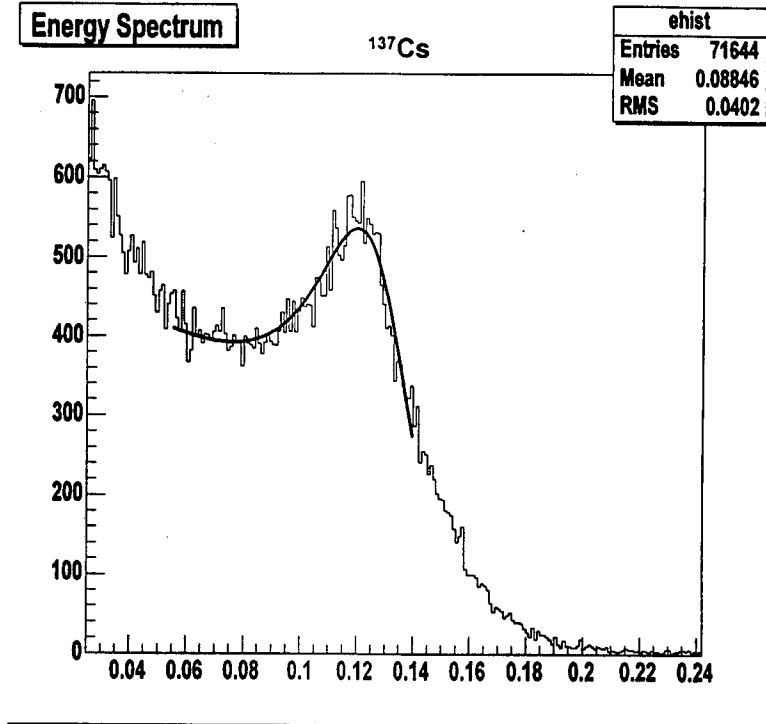


Figure 6. ^{137}Cs γ spectrum. A theoretical Compton edge spectrum smeared by a fixed detector resolution is fit to the data.

After background subtraction, the efficiency is determined by measuring the neutron rate above some proton recoil energy threshold and dividing by the total neutron yield for a given source. The neutron rate is determined by either PSD method described above. The γ contamination is defined as the fraction of neutrons that are really misidentified γ events. ^{60}Co source data is used to determine the ratio of misidentified γ events to properly identified γ events. This fraction is measured in 10 KeV energy bins for both PSD methods. It is then multiplied by the ratio of γ to neutron events identified by PSD in ^{252}Cf source data to determine the γ contamination. The γ contamination is subtracted from the efficiency. Table 1 shows the measured efficiency and γ contamination for a ^{252}Cf source. The Combined Algorithms method appears to give a lower γ contamination for comparable efficiency. Table 2 shows the efficiency for detecting AmLi neutrons for the Combined Algorithms method. Increasing the threshold can render a significant reduction in γ contamination and interrogation source neutrons with only a small sacrifice in efficiency.

Table 1. Efficiency and γ contamination for a range of electron and proton recoil energy thresholds. The Combined Algorithms PSD method gives a lower γ contamination for comparable efficiency compared to the χ^2 method.

Energy Threshold		Combined Algorithms		χ^2 Method	
E_e (KeV)	E_p (KeV)	Effic (%)	γ Cont (%)	Effic (%)	γ Cont (%)
40	530	2.66	1.04	2.79	1.9
50	580	2.58	0.75	2.65	1.54
60	620	2.46	0.52	2.52	1.23
70	660	2.34	0.38	2.38	0.99
80	690	2.22	0.31	2.26	0.85
90	720	2.11	0.23	2.14	0.68
100	750	2.01	0.18	2.04	0.54

Table 2. Efficiency for AmLi source data using only the Combined Algorithms PSD method. γ contamination is neither estimated nor subtracted.

Energy Threshold		
E_e (KeV)	E_e (KeV)	Efficiency (%)
40	40	0.61
50	50	0.54
60	60	0.46
70	70	0.37
80	80	0.29
90	90	0.24
100	100	0.19

III. Conclusions

The ability of this data acquisition and analysis technique to separate the neutrons from the gammas and to discriminate the fission neutrons from activation neutrons is very promising. The efficiencies measured for the single module indicate that an active well counter can be designed and built to give total efficiency of $\sim 25\%$. Such a device with coincidence resolving times on the order of tens of nanoseconds will provide an order of magnitude improvement in facility throughput or range of sample size.

Much work is yet to be done to make this a reality. The results of this study will be incorporated into a Monte Carlo simulation program. This program will be used to optimize the custom module design and configuration of a prototype counter. Several liquid scintillator modules will be procured and compared before choosing the best vendor to supply the optimized detector modules. Once a complete set of modules has been purchased and configured, we will quantify the response and efficiency of the counter. A set of electronics to handle event triggering, high voltage supply, and data acquisition will be purchased and integrated into the counter. Finally, measurements will be made with known standards and facility nuclear materials.

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